Localizing periodicity in near-field images

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We show that Bayesian inference, like that used in statistical mechanics, can guide the systematic construction of Fourier dark-field methods for localizing periodicity in near-field (e.g. scanning-tunneling and electron-phase-contrast) images. For crystals in an aperiodic field, the Fourier coefficient $Ze^{i\varphi}$ combines with a prior estimate for background amplitude B to predict background phase (β) values distributed with a probability $p(\beta - \varphi \mid Z, \varphi, B)$ inversely proportional to the amplitude P of the signal of interest, when this latter is treated as an unknown translation scaled to B. From UMStL-CME-90f26pf.

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I. INTRODUCTION

Near-field images (defined here as images of wave amplitude or phase at the exit surface of a solid) with atomic (i.e. less than 2Å) resolution are developments of the last quarter of this century. Transmission electron microscopes capable of delivering phase-contrast images with continuous transfer to spatial frequencies beyond $1/(2\text{Å})$ have become available in the last decade,[1] and the first scanning tunneling[2] and atomic-force[3] microscopes able to resolve atoms have been created in this period as well. More recently, near-field visible-light microscopes with resolutions much below the wavelength limit normally associated with light microscopes, although not with atomic resolution, have been described as well.[4]

As viewed from frequency space, near-field images with atomic resolution may contain data of three basic types. The first type, which we refer to as the "diffraction data", are simply the data on lattice periodicity amplitudes contained in image power spectra. All researchers accustomed to obtaining data on lattice parameters or orientation from diffraction patterns have experience with this information. Near-field images, sometimes under a more restricted set of conditions, can also contain "phase-information" on the phase-lag from one periodicity to the next. Diffractionists involved in structure determination will recognize that such information is needed, along with diffraction data, to determine the distribution of scattering density within unit cells. Finally, such images also contain "darkfield information", which we define here as information on amplitude and phase differences across the breadth of individual diffracted beams. This information tells how near-by periodicities interfere via "beat" processes, and hence how the intensity of any given range of periodicities is distributed throughout the region examined. In other words, it tells where crystals and their boundaries are located in the image field. Researchers involved in small-angle and anomalous Bragg-scattering studies, as well as in diffraction imaging techniques (like x-ray topography or weak-beam electron imaging), all use this sort of information. Electron and x-ray dark-field techniques which use far-field diffraction contrast to provide data on both phase and amplitude components of this information, in particular, have served for decades as powerful tools for the study of defects in (and boundaries around) crystals.[5]

Although the darkfield data in high-resolution near-field images are of demonstrable interest for complementing that available via the far-field techniques discussed above, methods for extracting that information have generally taken the form of qualitative and *ad hoc* "recipes" for image processing.[6] As one consequence of the large and growing amount of data in individual images (e.g., tens of megabytes available in individual electron-phase-contrast negatives), formal techniques of physical inference (like the theory of accessible state probabilities used in statistical mechanics) can facilitate more rigorous and quantitative study. In this paper, we outline a strategy for taking steps in this regard. With Bayes' theorem as our prescription for physical inference from new data,[7] we first tackle the problem of extracting darkfield information on periodic structures buried in an otherwise aperiodic field. This problem is common to electron-phase-contrast and air-based scanning-tunneling images, even of purely crystalline fields, because specimen preparation and system instabilities, respectively, often give rise to a superposed aperiodic background. Second, we discuss how the strategy may be extended to treat a wider class of problems as well.

II. THEORY

Mathematically, suppose we are given an experimental image $z[x, y]$ whose Fourier transform is $\widetilde{\mathbf{Z}}[u, v]$, and that the objective is to construct a darkfield image of selected regions of u,v space associated with one or more peaks in the power spectrum $Z^2[u, v]$. Although we will neglect contributions to the darkfield image from regions of u,v space outside of those selected, in the "peak regions" we must separate the $\widetilde{\mathbf{Z}}[u, v]$ coefficients into peak $\widetilde{\mathbf{P}}$ and background $\widetilde{\mathbf{B}}$ components such that $\widetilde{\mathbf{P}}[u, v] \equiv \widetilde{\mathbf{Z}}[u, v] - \widetilde{\mathbf{B}}[u, v]$. The "darkfield image" is then just the inverse transform of $\widetilde{\mathbf{P}}$, namely $p[x, y] = z[x, y] - b[x, y]$. Here **bold** print denotes a complex number. We have assumed also that the starting image is real, and that $\widetilde{\mathbf{P}}$ and $\widetilde{\mathbf{B}}$ as chosen retain the conjugate symmetry of $\widetilde{\mathbf{Z}}$ (i.e. $\widetilde{\mathbf{Z}}[-u, -v] = \widetilde{\mathbf{Z}}^*[u, v]$) so that their inverse transforms are real as well.

In order to separate $\widetilde{\mathbf{Z}}$ into peak and background components, information on the nature of the background is crucial. Previously proposed methods make *ad hoc* assumptions about $\widetilde{\mathbf{B}}[u, v]$. For example, the traditional "window method" assumes that $\widetilde{\mathbf{B}}[u, v]$ is zero inside the peak regions. The usefulness by comparison of taking explicit account of estimates for the background amplitude beneath peaks was recently demonstrated by O'Keefe and Sattler, but only *ad hoc* assumptions (such as a uniformly random distribution) were proposed for the assignment of background phases.[8] If, however, information on background phase is taken into account systematically, noise in the calculations can be reduced further, and the background-subtraction recipe can serve as a basis for a plethora of strategies for direct physical inference.

Specifically, if for any value of $[u, v]$ we consider calculation of the posterior probability $p(\beta \mid \widetilde{\mathbf{Z}}, B)d\beta$ of the background phase β, given the measured coefficient $\widetilde{\mathbf{Z}} \equiv Ze^{i\varphi}$ and in addition a prior information value for the background amplitude B, then Bayes' theorem gives

$$p(\beta \mid Z, \varphi, B)d\beta = p(\beta \mid Z, B)d\beta \frac{p(\varphi \mid Z, B, \beta)}{p(\varphi \mid Z, B)}. \tag{1}$$

Equation (1) tells us how to modify our estimate of the background phase β in light of data on the phase φ of $\widetilde{\mathbf{Z}} = \widetilde{\mathbf{P}} + \widetilde{\mathbf{B}}$ at that frequency. In notational terms, $p(\beta \mid Z, B)d\beta$ is the *prior probability* of β given Z and B; $p(\varphi \mid Z, B, \beta)d\varphi$ is the *likelihood function* (or sampling distribution) probability that φ will lie between φ and $\varphi + d\varphi$ for given values of Z, B, and β; and $p(\varphi \mid Z, B)d\varphi$ is the *a priori probability* of φ given Z and B.

The prior $p(\varphi \mid Z, B)d\varphi$ will of course have no β dependence, since it can be considered an integral over all β. Determination of such β-independent terms can be considered academic here, since $p(\beta \mid Z, B)d\beta$ obeys the normalization condition

$$\int_0^{2\pi} p(\beta \mid Z, B)d\beta = 1. \tag{2}$$

The β dependence of $p(\beta \mid Z, B)d\beta$, one of the fundamental priors, will be held for discussion below. the β dependence of $p(\varphi \mid Z, B, \beta)d\varphi$, on the other hand, is less fundamental, and is complicated by the fact that $\widetilde{\mathbf{Z}}$ is a composite of two physically distinct quantities ($\widetilde{\mathbf{P}}$ and $\widetilde{\mathbf{B}}$). Fortunately, we can replace this φ probability with the product of a probability for α, the peak phase angle, and a geometric term. This is done by writing the peak phase angle α in terms of Z, φ, B, and β, and then changing differential variables in the normalization equation for $p(\alpha \mid Z, B, \beta)d\alpha$, to get

$$p(\varphi \mid Z, B, \beta)d\varphi = p(\alpha \mid Z, B, \beta) \left| \frac{\partial \alpha}{\partial \varphi} \right|_{Z, B, \beta} d\varphi. \tag{3}$$

Here the second factor on the right-hand side is the absolute value of the Jacobian (functional determinant) for the variable change.

To further eliminate $\widetilde{\mathbf{Z}}$ dependences, note from Bayes' theorem that

$$p(\alpha \mid Z, B, \beta)d\alpha = p(\alpha \mid B, \beta)d\alpha \frac{p(Z \mid \alpha, B, \beta)}{p(Z \mid B, \beta)}. \tag{4}$$

As above, a variable change (here from Z to P) then allows us to write

$$p(Z \mid \alpha, B, \beta)dZ = p(P \mid \alpha, B, \beta) \left| \frac{\partial P}{\partial Z} \right|_{\alpha, B, \beta} dZ. \tag{5}$$

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The partials in (3) and (5) are easily calculated, and hence these expressions allow us to evaluate the probability $p(\beta \mid Z, \varphi, B)d\beta$ for *any* assignment of the peak and background prior probabilities $p(\beta \mid Z, B)d\beta$, $p(\alpha \mid B, \beta)d\alpha$, $p(P \mid \alpha, B, \beta)dP$.

To evaluate the partials, first note from Fig. 1 that Z can be written in terms of P, α, B, and β as

$$Z = \left\{ (P\cos[\alpha] + B\cos[\beta])^2 + (P\sin[\alpha] + B\sin[\beta])^2 \right\}^{\frac{1}{2}}, \tag{6}$$

and that α can be related to the variables Z, φ, B, and β via the expressions

$$\varphi - \alpha = \arctan\left[\frac{B\sin[\beta - \varphi]}{B\cos[\beta - \varphi] - Z} \right] \tag{7}$$

$$= \arccos\left[\frac{B\cos[\beta - \varphi] - Z}{B^2 - 2ZB + Z^2} \right].$$

At this point, it is convenient to introduce the following notation for signed angles in the triangle formed by $\widetilde{\mathbf{Z}}$, $\widetilde{\mathbf{P}}$, and $\widetilde{\mathbf{B}}$: $\Theta_B \equiv \varphi - \alpha$, $\Theta_P \equiv \beta - \varphi$, and $\Theta_Z \equiv \alpha - \beta - \pi\times\text{sgn}[\alpha - \beta]$, where $\text{sgn}[x] \equiv \{+1 \text{ for } x \geq 0, -1 \text{ for } x < 0\}$. Note also the mathematical equivalence of Θ_Z and Θ_B under exchange of $\widetilde{\mathbf{Z}}$ and $\widetilde{\mathbf{B}}$.

By taking the derivative of (6) with respect to P at constant α, B, and β, it is easy to show that $(\partial Z/\partial P)_{\alpha,B,\beta} = \widetilde{\mathbf{Z}} \bullet \widetilde{\mathbf{P}}/ZP = \cos[\Theta_B]$, and hence $|\partial P/\partial Z|_{\alpha,B,\beta} = |\sec[\Theta_B]|$. By differentiating (7) with respect to φ at constant Z, B, and β, it likewise follows that

$$\left| \frac{\partial \alpha}{\partial \varphi} \right|_{Z,B,\beta} = \left| \frac{1 - (B/Z)\cos[\beta - \varphi]}{(B/Z)^2 - 2(B/Z)\cos[\beta - \varphi] + 1} \right| \tag{8}$$

$$= \frac{|\cos[\Theta_B]|}{P/B}.$$

Note, therefore, that the product of the two partials is simply B/P. These facts in hand, we can turn to assignment for the prior probabilities $p(\beta \mid Z, B)d\beta$, $p(\alpha \mid B, \beta)d\alpha$, and $p(P \mid \alpha, B, \beta)dP$ mentioned above.

For the frequent times when prior information singles out no preferred direction, uniform (isotropic) priors for both β and α [i.e., $p(\beta \mid Z, B) = p(\alpha \mid B, \beta) = 1/2\pi$] will be appropriate. In any case, these priors do not enter into other potentially β-dependent terms in Eqs. (1)-(5). Hence, should we assign them an explicit β-dependence it will be factored into $p(\beta \mid Z, \varphi, \beta)$ directly.

The most complicated prior is $p(P \mid \alpha, B, \beta)$, because it cannot be dismissed by isotropy, and because it enters also into the only term whose β-dependence remains undiscussed, namely $p(Z \mid B, \beta)$ in Eq.(4). We point out here two choices of potential interest. A prior for P which (i) appears simple to implement, (ii) results in correlations as expected between the direction of B and Z when $B \approx Z$, but (iii) has unclear physical meaning at best is that which makes $p(\alpha \mid Z, B, \beta)$ in Eq. (3) a constant. The probability distribution for β resulting from such a prior is plotted in Fig. 2. To generate pseudorandom values for β with this distribution, one can simply choose values of Θ_Z (hence α) uniformly distributed in its allowed range for given Z/B, and then calculate $\beta - \varphi = \Theta_P$ from Θ_Z allowing for the fact that Θ_P is a double-valued function of Θ_Z with 50% of its values on each branch when $Z < B$. Numerical simulations in our laboratory have verified that a pseudorandom-number generator so constructed reproduces the distribution shown in Fig. 2.

The physically (if not computationally) most helpful assignment for $p(P \mid \alpha, B, \beta)$ is likely to be the uniform (β-independent) assignment. If we view P not as a scale factor but as the magnitude of a vector translation whose length is scaled to the value of B, then $p(P \mid \alpha, B, \beta) = const = 1/P_{\max}$ for $P_{\max} \gg Z_{\max}$ may be considered the correct transformation-invariant prior probability in the absence of further information.[9] In this case,

$$p(Z \mid B, \beta) = \frac{1}{P_{\max}} \int_0^{2\pi} |\sec[\Theta_B]|\, d\Theta_Z. \tag{9}$$

The argument in this integral is double-valued and must therefore be handled carefully for $\Theta_B < \Theta_Z$, but it can nevertheless be written as a function of Z, B, and Θ_Z only. Hence the integral itself is independent of β. Given this uniform prior for P, and isotropic priors for α and β, Eqs. (1)-(5) therefore give a β-dependence for $p(\beta \mid Z, \varphi, B)$ proportional to the product of partials, and inversely proportional to P. This distribution is plotted for various Z/B in Fig. 3. Numerical simulations in our laboratory have confirmed that P vectors with a uniform distribution of lengths and orientation angles yield β-values with the distribution shown.

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III. SUMMARY

The Bayesian phase model here does two things. First of all, it takes the image-processing recipe for "background subtraction" proposed by O'Keefe and Sattler and creates a recipe for physical inference by replacing *ad hoc* assignments of background phase with an assignment of phases based on physical assumption. Second, it systematically minimizes noise in darkfield images associated with crystallite fine structure (e.g., boundaries), since the information on fine structure is found in outlying regions of the Bragg peaks where Z/B is near 1. These are the regions where errors associated with an *ad hoc* phase assignment will have their greatest effect. Because the image itself, not the imaging process, constitutes the object of the physics in this analysis, it can be applied to images inferred via Bayesian statistical analysis of the imaging process[10] as well as to raw experimental data.

Strategies for obtaining prior information on the amplitude of B in these applications begin with simple extrapolation of an azimuthally symmetric "cloud" of aperiodic contrast in Fourier space.[11] Applications of Fourier darkfield analysis which are now limited by resolution and signal-to-noise ratio include the study of isotopically anomalous nm-sized diamonds in meteorites,[12] and the study of objects showing space-group disallowed symmetries due to twinning and quasicrystallinity. The algorithms here facilitate removal of aperiodic background from selected frequency-space regions in images of these structures. Some applications, such as the study of icosahedral structures in periodic array,[13] may require removal of periodic background. These involve prior information which goes beyond the scope of the result, but not the formalism, discussed here.

The formalism is also not limited to prior information on background amplitude. Prior information on the variance in background amplitude (here assumed to be zero) is a logical addition. When it can be made a practical addition remains to be seen. The formalism should also allow us to estimate the strength of periodicities (analogous to diffracted beam intensities) from point to point in the field. This is something that diffraction darkfield does which Fourier darkfield does less directly. However, the strategy posed here for physical inference from images allows this and other questions to be asked mathematically. As another example, diffraction darkfield imaging does not allow us to distinguish between crystals with the same periodicity but a different periodicity phase. Atomic-resolution images with data on periodicity phase and amplitude contain the needed data, and the strategy proposed here suggests a formalism for posing this question as well.

[1] Compare J. C. Spence, *Experimental High-Resolution Electron Microscopy* (Oxford Univ. Press, New York, 1981 and 1988), 1st and 2nd ed.

[2] G. Binnig and H. Rohrer, Helv. Phys. Acta **55**, 762 (1982).

[3] G. Binnig, C. F. Quate, and Ch. Gerber, Phys. Rev. Lett. **56**, 930 (1986).

[4] E. Betzig, M. Isaacson, and A. Lewis, Appl. Phys. Lett. **51**, 2088 (1987); U. Durig, D. W. Pohl, and F. Rohner, Appl. Phys. **59**, 3318 (1986); M. Isaacson, in *Proceedings of the Forty-Seventh Annual Meeting of the Electron Microscopy Society of America*, edited by G. W. Bailey (San Francisco Press, San Francisco, 1989), p. 4.

[5] Compare P. B. Hirsch, A. Howie, R. B. Nicholson, D. W. Pashley, and M. J. Whelan, *Electron Microscopy of Thin Crystals* (Butterworths, London, 1965).

[6] P. R. Buseck, Y. Epelboin, and A. Rimsky, Acta Crystallogr. Sect. A **44**, 975 (1988); R. Perez and J. g. Perez-Ramirez, in *Proceedings of the Forty-Sixth Annual Meeting of the Electron Microscopy Society of America*, edited by G. W. Bailey (San Francisco Press, San Francisco, CA, 1988), p. 838.

[7] E.g. W. Grandy, *Statistical Mechanics Vol. I* (Reidel, Dordrecht, 1987).

[8] M. A. O'Keefe and M. L. Sattler, in *Proceedings of the Forty-Fifth Annual Meeting of the Electron Microscopy Society of America*, edited by G. W. Bailey (San Francisco Press, San Francisco, CA, 1987), p. 104; in *Proceedings of the Forty-Sixth Annual Meeting of the Electron Microscopy Society of America* (Ref. 6), p. 834.

[9] E. T. Jaynes, IEEE Trans. Systems Sci. Cybernet **4**, 227 (1968).

[10] For example, E. J. Kirkland, in *Scanning Microscopy Supplement 2* (Scanning Microscopy International, Chicago, 1988), p. 139.

[11] P. Fraundorf and G. K. Fraundorf, in *Proceedings of the Forty-Seventh Annual Meeting of the Electron Microscopy Society of America* (Ref. 4), p. 122; Monsanto Report No. MSL-9347 (Monsanto Information Services, St. Louis, 1989); P. Fraundorf and J. Tentschert, in Abstracts to the Thirty-Seventh Midwest Solid State Conference, Rolla, Missouri, 1989 [Appl. Phys. Commun. (to be published)].

[12] P. Fraundorf, G. K. Fraundorf, T. Bernatowicz, R. S. Lewis, and T. Ming, Ultramicroscopy **27**, 401 (1989); M. A. O'Keefe, D. Blake, F. Freund, C. Hetherington, and J. Turner, in *Proceedings of the Forty-Sixth Annual Meeting of the Electron Microscopy Society of America* (Ref. 4), p. 836.

[13] L. Levine, K. Kelton, and P. Gibbons, in Abstracts to the Thirty-Seventh Midwest Solid State Conference (Ref. 11).

FIG. 1. Given complex Fourier coefficients obeying $Ze^{i\varphi} = Pe^{i\alpha} + Be^{i\beta}$, this schematic illustrates the geometric relationship between peak phase angle α, and background phase angle β, when Z, φ, and B are specified.

FIG. 2. A plot of $p(\beta \mid Z, \varphi, B)$ in probability per radian interval vs. $\beta - \varphi$ in radians for the computationally convenient case when $p(\beta \mid Z, B) = 1/2\pi$, and $p(\alpha \mid Z, B, \beta)$ is independent of β. Note the strong bias toward $\beta \approx \varphi$ as Z/B approaches 1, and the cusps for $Z/B < 1$. An algorithm for generating β values with this distribution is described in the text. Notation for Z/B: solid line, $\frac{1}{8}$; plus, $\frac{1}{2}$; cross, 2; diamond, 8.

FIG. 3. A plot of $p(\beta \mid Z, \varphi, B)$ in probability per radian interval vs. $\beta - \varphi$ in radians for the physically realistic case when $p(\beta \mid Z, B) = p(\alpha \mid B, \beta) = 1/2\pi$, and $p(P \mid \alpha, B, \beta) = const = 1/P_{\max}$, where $P_{\max} \gg Z_{\max}$. Note the bias toward $\beta \approx \varphi$ as Z/B approaches 1. The bias will weaken with increasing variance in B when nonzero variances are included as prior information in the calculation. Notation for Z/B: solid line, $\{512, \frac{1}{512}\}$; plus, $\{8, \frac{1}{8}\}$; cross, $\{2, \frac{1}{2}\}$; diamond, $\{2^{1/3}, 2^{-1/3}\}$.

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